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MERGING HYPERSPECTRAL IMAGERY AND MULTI-SCALE MODELING FOR LASER LETHALITY

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14. ABSTRACT
The overall goal of the project was to develop advanced computational models for analysis of the effect of external flow on material removal in laser ablation of metals in oxidizing environment and under the influence of air flow. The models are applied for investigation of the combined effect of the laser energy deposition, chemical reactions, and air flow on the material removal rate and the nature of laser damage/modification of irradiated targets. The results of this project include the elucidation of (1) the relative contributions of the recoil vapor pressure and Marangoni effects on the melt dynamics and meltthrough time in the laser melting of free-standing aluminum films, (2) the effect of the external gas pressure on the flow structures and the mechanisms of the alumina and oxygen transport to the target surface, (3) the distinct characteristics of short pulse laser interactions with a metal target under conditions of spatial confinement by a solid transparent overlayer, (4) the conditions for the formation of nanocrystalline surface layer, sub-surface voids, and frozen surface nanospikes in surface nanostructuring by short laser pulses, (5) the effect of sample porosity on laser-induced carbon pyrolysis and oxidation

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Merging Hyperspectral Imagery and Multi-Scale Modeling for Laser Lethality

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Dates of contract period: 09.15.10 – 09.14.2015

This report covers the part of the research program conducted at the University of Virginia and University of Alabama.

Technical Abstract:

The overall goal of the project is to develop advanced computational models for analysis of the effect of external flow on material removal in laser ablation of metals in oxidizing environment and under the influence of air flow. A multi-scale computational approach combining several computational methods, from atomistic to continuum, is adopted for investigation of different aspects of the ablation process with different spatial resolutions and different level of accuracy. The main focus of the computational effort is on providing the physical understanding of the combined effect of the laser energy deposition, chemical reactions, and air flow on the material removal rate and the nature of laser damage/modification of the irradiated target. Some of the key results of this project are as follows.

(1) Carbon pyrolysis and oxidation model is developed. The model includes an accurate description of the surface interface conditions with heat and mass transfer. The carbon pyrolysis and oxidation model are applied for analysis of laser heating effects (surface temperature) and flow velocity, as well as the effect of sample porosity elucidated using porous and non-porous heterogeneous kinetic approaches.

(2) An advanced computational model for two-phase hydrodynamic simulations of continuous wave (CW) laser interactions with metals is developed and used for investigation of the relative contributions of the recoil vapor pressure and Marangoni effects on the melt dynamics and melt-through time in the laser melting of free-standing aluminum films. A series of simulations performed with this model reveal the conditions when the Al melt expulsion is dominated by either vapor pressure recoil or Marangoni effect.

(3) A multi-phase computational model for the CW laser heating, melting, and vaporization of Al films with air flow is developed. The direct simulation Monte Carlo method enhanced with a simplified description of Al vapor burning in air is used in the simulations. Vapor expansion from Al target irradiated by a continuous wave laser into a supersonic external air flow is investigated in kinetic simulations performed for a broad range of pressure in the external flow. The results of the simulations reveal a significant effect of the external gas pressure on the flow structures and the mechanisms of the alumina and oxygen transport to the target surface.

(4) New methods for incorporation of the description of a transparent overlayer (or an oxide layer) into a hybrid atomistic – continuum computational model is developed and applied for investigation of the distinct characteristics of short pulse laser interactions with a metal target under conditions of spatial confinement by a solid transparent overlayer.

(5) Surface nanostructuring by short laser pulses is investigated in the irradiation regime close to the ablation/spallation thresholds. The conditions for the formation of nanocrystalline surface layer, sub-surface voids (surface swelling), and frozen surface nanospikes with unusual polyicosahedral nanostructure are elucidated.

Publications:

1. A. N. Volkov and L. V. Zhigilei, “Computational study of the role of oxidation in continuous wave laser ablation of an aluminum target into an external supersonic air flow,” Appl. Phys. A 110, 537–546, 2013.
2. C. Wu and L. V. Zhigilei, Microscopic mechanisms of laser spallation and ablation of metal targets from large-scale molecular dynamics simulations, *Appl. Phys. A* **114**, 11-32, 2014.
3. C. Wu, E. T. Karim, A. N. Volkov, and L. V. Zhigilei, Atomic movies of laser-induced structural and phase transformations from molecular dynamics simulations, in *Lasers in Materials Science*, Springer Series in Materials Science, Vol. **191**, Edited by M. Castillejo, P. M. Ossi, and L. V. Zhigilei (Springer International Publishing Switzerland, 2014), pp. 67-100.
4. E. T. Karim, M. Shugaev, C. Wu, Z. Lin, R. F. Hainsey, and L. V. Zhigilei, Atomistic simulation study of short pulse laser interactions with a metal target under conditions of spatial confinement by a transparent overlayer, *J. Appl. Phys.* **115**, 183501, 2014.
5. C. M. Rouleau, C.-Y. Shih, C. Wu, L. V. Zhigilei, A. A. Puretzky, and D. B. Geohegan, Nanoparticle generation and transport resulting from femtosecond laser ablation of ultrathin metal films: Time-resolved measurements and molecular dynamics simulations, *Appl. Phys. Lett.* **104**, 193106, 2014.
6. E. T. Karim, C. Wu, and L. V. Zhigilei, Molecular dynamics simulations of laser-materials interactions: General and material-specific mechanisms of material removal and generation of crystal defects, in *Fundamentals of Laser-Assisted Micro- and Nanotechnologies*, Springer Series in Materials Science, Vol. **195**, Edited by V. P. Veiko and V. I. Konov (Springer International Publishing Switzerland, 2014), pp. 27-49.
7. C. Wu, M. S. Christensen, J.-M. Savolainen, P. Balling, and L. V. Zhigilei, Generation of sub-surface voids and a nanocrystalline surface layer in femtosecond laser irradiation of a single crystal Ag target, *Phys. Rev. B* **91**, 035413, 2015.
8. C. Wu and L. V. Zhigilei, Nanocrystalline and polyicosahedral structure of a nanospike generated on metal surface irradiated by a single femtosecond laser pulse, *J. Phys. Chem. C*, in press, 2016, DOI: 10.1021/acs.jpcc.6b00013
9. E. T. Karim, M. V. Shugaev, C. Wu, Z. Lin, H. Matsumoto, M. Conneran, J. Kleinert, R. F. Hainsey, and L. V. Zhigilei, Experimental characterization and atomistic modeling of

interfacial void formation and detachment in short pulse laser-processing of metal surfaces covered by transparent overlayers, *Appl. Phys. A*, in press.

10. A. N. Volkov and L. V. Zhigilei, Melt dynamics and melt-through time in continuous wave laser heating of metal films: Contributions of the recoil vapor pressure and Marangoni effects, *Int. J. Heat Mass Transfer*, to be submitted, 2016, draft of the paper is available upon request.
11. R. F. Johnson and H. K. Chelliah, Laminar reactive boundary layer simulation of an ablating heated carbon surface, *51st AIAA Aerospace Sciences Meeting including the New Horizons Forum and Aerospace Exposition*, 184, 2013.
12. R. F. Johnson, A. C. VanDine, G. Esposito, and H. K. Chelliah, On the axisymmetric counterflow flame simulations: Is there an optimal nozzle diameter and separation distance to apply quasi one-dimensional theory?, *Combustion Science and Technology* **187,** 37-59, 2015.
13. R. F. Johnson and H. K. Chelliah, Numerical simulation of two-dimensional flow over a heated carbon surface with coupled heterogeneous and homogeneous reactions, accepted for publication in *Combustion Theory and Modeling*, 2016, in print.
14. R. F. Johnson and H. K. Chelliah, Coupled heterogeneous and homogeneous reactions in a two-dimensional stagnation-point flow field, to be submitted *Combustion Theory and Modeling, 2016.*

Presentations:

L. V. Zhigilei, (Plenary Talk) Computer modeling of material modification by short laser pulses and optically-induced surface acoustic waves, Progress in Electromagnetics Research Symposium (PIERS 2015), Prague, Czech Republic, July 06-09, 2015.

L. V. Zhigilei, Atomistic modeling of material modification by femtosecond laser pulses and surface acoustic waves, Physics Department Colloquium Series at Virginia Commonwealth University, Richmond, Virginia, April 17, 2015.

L. V. Zhigilei, (Invited Talk) Atomistic modeling of material modification by femtosecond laser pulses and surface acoustic waves, 6th European Conference on Applications of Femtosecond Lasers in Materials Science (FemtoMat), Mauterndorf / Salzburg, Austria, March 16-17, 2015.

L. V. Zhigilei, C. Wu, M. V. Shugaev, and E. T. Karim, Large-scale atomistic modeling of structural modification of metal surfaces in femtosecond laser processing, 13th Conference on Laser Ablation (COLA-2015), Cairns, Australia, 31 August – 4 September, 2015.

L. V. Zhigilei, C.-Y. Shih, C. Wu, and M. V. Shugaev, Molecular dynamics simulation study of femtosecond laser ablation of silver thin films and bulk targets in water environment, 13th Conference on Laser Ablation (COLA-2015), Cairns, Australia, 31 August – 4 September, 2015.

M. V. Shugaev, E. T. Karim, C.-Y. Shih, C. Wu, and L. V. Zhigilei, Interaction of short pulse laser irradiation with metal targets under condition of spatial confinement, APS March Meeting, San Antonio, Texas, March 2 – 6, 2015.

C. Wu, M. Shugaev, and L. V. Zhigilei, Large-scale atomistic simulations of surface nanostructuring by short pulse laser irradiation, APS March Meeting, San Antonio, Texas, March 2 – 6, 2015.

A. N. Volkov, (Invited Talk) Melt dynamics and melt-through time in continuous wave laser ablation: Comparative analysis of contributions from the recoil vapor pressure and Marangoni effect, International

Symposia on High-Power Laser Ablation and Beamed Energy Propulsion, Santa Fe, New Mexico, April 21-25, 2014.

L. V. Zhigilei, (Invited Talk) Large-scale atomistic simulations of the phase transformations and surface modification in short pulse laser processing of metals, The Institute of Optics, University of Rochester, Rochester, New York, December 1, 2014.

L. V. Zhigilei, (Invited Talk) Large-scale atomistic simulations of the structural transformations and microstructure development in short pulse laser processing of metals, Laser Processing and Fabrication for Solar, Displays, and Optoelectronic Devices III, SPIE Optics + Photonics 2014, San Diego, California, August 17-21, 2014.

L. V. Zhigilei, (Tutorial Lecture) Atomistic and coarse-grained molecular dynamics simulations of laser-materials interactions, 4th Venice International School on Lasers in Materials Science, Isola di San Servolo, Venice, Italy, July 13-20, 2014.

L. V. Zhigilei, (Invited Talk) Generation of nanoparticles in short pulse laser ablation of bulk metal targets and thin films: Insights from large-scale atomistic simulations, 15th International Symposium on Laser Precision Microfabrication (LPM 2014), Vilnius, Lithuania, June 17-20, 2014.

L. V. Zhigilei, (Invited Talk) Large-scale atomistic simulations of the structural transformations and microstructure development in short pulse laser processing of metals, European Materials Research Society (E-MRS) 2014 Spring Meeting, Lille, France, May 26-30, 2014.

A. N. Volkov and L. V. Zhigilei, Multiphase modeling of continuous wave laser ablation: Contributions of the recoil vapor pressure, Marangoni effect, and external gas flow, ASME 2014 International Mechanical Engineering Congress & Exposition, Montreal, Canada, November 14-20, 2014.

C. Wu, E. T. Karim, M. Shugaev, C.-Y. Shih, and L. V. Zhigilei, Large-scale atomistic simulations of laser ablation, generation of sub-surface voids, crystal defects and nanocrystalline surface layers in short pulse laser processing of metals, 9th International Conference on Photo-Excited Processes and Applications, Matsue, Japan, September 29 - October 3, 2014.

C.-Y. Shih, C. Wu, M. Shugaev, L. V. Zhigilei, C. M. Rouleau, A. A. Puretzky, and D. Geohegan, Atomistic simulations and experimental study of nanoparticle generation in femtosecond laser ablation of thin metal films, International Symposia on High-Power Laser Ablation and Beamed Energy Propulsion, Santa Fe, New Mexico, April 21-25, 2014. (First Place Award)

E. T. Karim, M. Shugaev, C. Wu, C.-Y. Shih, L. V. Zhigilei, Z. Lin, and R. Hainsey, Atomistic simulation study of short pulse laser interactions with metal targets under conditions of spatial confinement, International Symposia on High-Power Laser Ablation and Beamed Energy Propulsion, Santa Fe, New Mexico, April 21-25, 2014.

C. Wu and L. V. Zhigilei, Large-scale massively parallel atomistic simulations of short pulse laser interaction with metals, APS 2014 March Meeting, Denver, Colorado, March 3-7, 2014.

C. M. Rouleau, A. A. Puretzky, D. B. Geohegan, M. Yoon, K. L. More, G. Duscher, C.-Y. Shih, C. Wu, and L. V. Zhigilei, Catalytic nanoparticles for carbon nanotube growth synthesized by through thin film femtosecond laser ablation, SPIE Photonics West 2014, San Francisco, California, February 1 – 6, 2014.

A. N. Volkov, (Invited Talk) Melt expulsion in continuous wave laser ablation: Contributions of the recoil vapor pressure, Marangoni effect, and external gas flow, International Symposium on Fundamentals of Laser Assisted Micro– and Nanotechnologies, St. Petersburg, Russia, June 24-28, 2013.

L. V. Zhigilei, (Invited Talk) Cooling rates and mechanisms of resolidification in short pulse laser processing of metal targets, 12th U.S. National Congress on Computational Mechanics, Raleigh, North Carolina, July 22-25, 2013.

L. V. Zhigilei, (Plenary Talk) Atomistic modeling of generation of crystal defects and microstructure development in short pulse laser processing of metals, International Symposium on Fundamentals of Laser Assisted Micro– and Nanotechnologies, St. Petersburg, Russia, June 24-28, 2013.

L. V. Zhigilei, (Invited Talk) Atomistic modeling of short pulse laser material modification, 1st Workshop on Materials in Extreme Environments (MatX), Michigan State University, East Lansing, Michigan, May 13-14, 2013.

R. F. Johnson, H. K. Chelliah, Laminar reactive boundary layer simulation of an ablating heated carbon surface, 51st AIAA Aerospace Sciences Meeting, Orlando, FL, January 2013.

R. F. Johnson, H. K. Chelliah, Coupled heterogeneous and homogeneous oxidation of heated carbon surfaces simulated using the OpenFOAM computational package, Combustion Institute ESS Meeting, Clemson, SC, October 2013.

R. F. Johnson, H. K. Chelliah, Numerical simulation of two-dimensional flow over a heated carbon surface with coupled heterogeneous and homogeneous reactions, 6th Mediterranean Combustion Meeting, Rhodes, Greece, June 2015.

Summary of some of the research findings and deliverables:

(1) ***Computational study of the role of gas-phase oxidation in CW laser ablation of Al target in an external supersonic air flow.*** Vapor expansion from Al target irradiated by a continuous wave laser into a supersonic external air flow is investigated in kinetic simulations performed for values of pressure in the external flow ranging from 50 Pa to 10000 Pa. The direct simulation Monte Carlo method enhanced with a simplified description of Al vapor burning in air is used in the simulations. The results of the simulations reveal a significant effect of the external gas pressure on the flow structures and the mechanisms of the alumina and oxygen transport to the target surface. At small values of pressure, the transport of both alumina and oxygen is controlled by diffusion and the formation of alumina film on the surface of the laser spot is dominated by the direct sedimentation of alumina from the gas flow. In this regime, the flux of oxygen to the laser spot is by an order of magnitude smaller than that of alumina. At higher values of pressure, the diffusion cannot directly deliver oxygen and alumina to the surface of the laser spot, but the circulating flow generated upstream the spot can effectively trap alumina formed in the flame zone and deposit it to the target surface. In this regime, the likely mechanism of the alumina film formation within the spot is the heterogeneous oxidation and growth of the film from the upstream edge of the spot in the downstream direction.

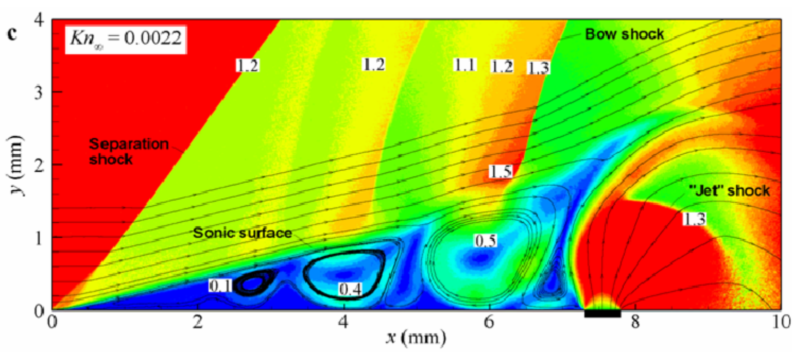


Figure 1: Mach number field in a simulation of the expansion and burning of Al vapor in the external air flow.

A. N. Volkov and L. V. Zhigilei, *Appl. Phys. A* **110**, 537, 2013.

(2) ***Melt dynamics and melt-through time in continuous wave laser heating of metal films: Contributions of the recoil vapor pressure and Marangoni effects.***

A numerical method based on the pseudo-compressibility formulation and upwind differencing is developed for two-phase hydrodynamic simulations of CW laser melting of metal films. This method is used to study the relative contributions of the recoil vapor pressure and Marangoni effects on the melt dynamics and melt-through time in the laser melting of free-standing aluminum films. The hydrodynamic simulations in the range of the film thickness from 0.2 mm to 4 mm and laser spot radius from 0.1 mm to 1 cm reveal only a marginal effect of the Marangoni stresses on the overall picture of the melt flow and melt-through time. The recoil effect, on the contrary, is found to be capable of sharply decreasing the melt-through time in a certain range of laser intensities. Below that range, the melting is dominated by the radial thermal transfer. Above that range, an active evaporation from the irradiated surface reduces the effectiveness of the melt expulsion. The range of laser intensities, where the melting is dominated by the recoil effect is not uniquely defined by the laser intensity and depends on the film thickness. A simple two-phase one-dimensional thermal model of laser melting, where the expulsion velocity due to the recoil effect is accounted for based on the Bernoulli integral, is developed to predict the melt-through time. This model is capable of capturing all qualitative trends revealed in the direct hydrodynamic simulations and accurately predicts the melt-through time above certain threshold laser intensity. The one-dimensional model can be used a robust engineering tool for the first-order estimates of the CW laser damage of metal films.

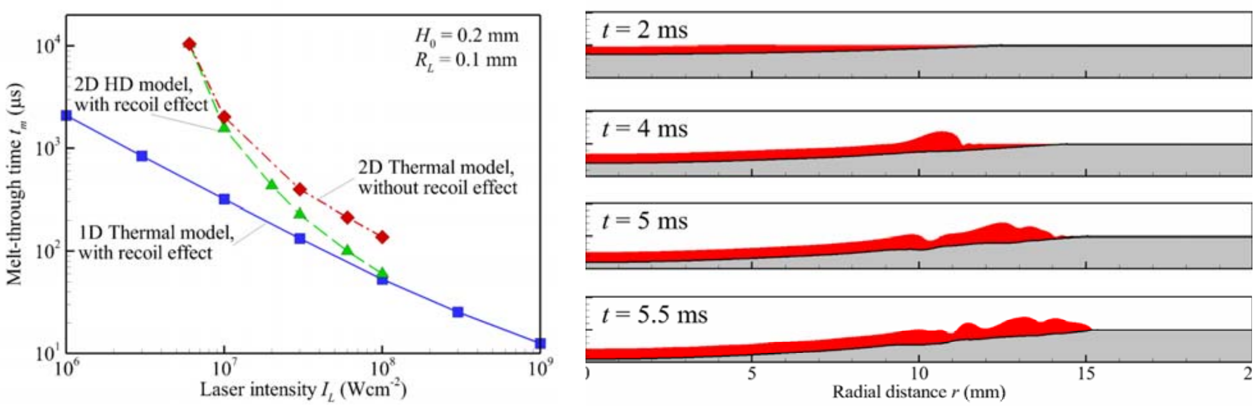
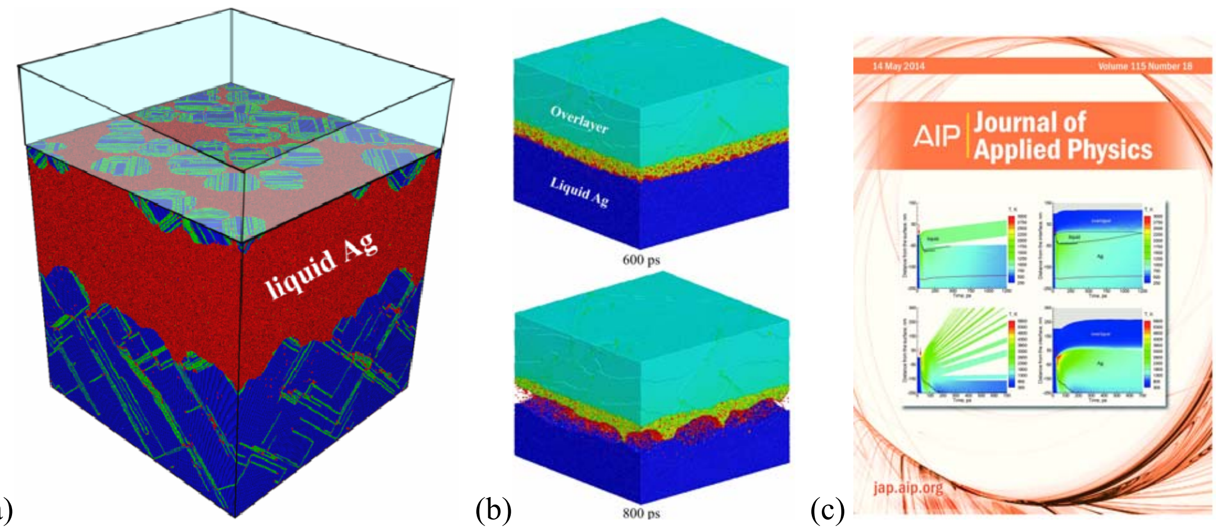


Figure 2: (left) Melt-through time versus laser intensity predicted with different computational models for an Al film with thickness of 0.2 mm and FWHM spot radius of 0.1 mm. (right) The shape of the molten pool (red) during laser melting of Al film of 1 mm thickness with the CW laser of intensity of 3×10^6 Wcm^{-2} and FWHM laser spot radius of 1 cm.

A. N. Volkov and L. V. Zhigilei, *Int. J. Heat Mass Transfer*, to be submitted, 2016, draft of the paper is available upon request.

(3) ***Atomistic simulation study of short pulse laser interactions with a metal target under conditions of spatial confinement by a transparent overlayer***. New methods for incorporation of the description of a transparent overlayer (or an oxide layer) into a hybrid atomistic – continuum computational model is developed and applied for investigation of the distinct characteristics of short pulse laser interactions with a metal target under conditions of spatial confinement by a solid transparent overlayer. The mechanisms responsible for structural modification of the metal-overlayer interfacial regions are investigated and processes responsible

for the generation of extended interfacial voids with internal nanoscale surface roughness, observed in experiments, are revealed. The results of the simulations demonstrate that the nucleation and growth of the interfacial voids are driven by the dynamic relaxation of laser-induced stresses proceeding simultaneously with rapid phase transformations and temperature variation in the interfacial region. The growth and coalescence of the interfacial voids result in the formation of liquid bridges connecting the overlayer and the metal substrate, whereas solidification of the transient liquid structures produced by breakup of the liquid bridges may be responsible for the formation of the nanoscale roughness of the interfacial voids observed in experiments. Computational analysis of the effect of pre-existing interfacial voids reveals a complex dynamic picture of the initial expansion and subsequent compaction of the surface region of the metal substrate and suggests a possible scenario for the formation of voids below the metal-overlayer interface



(a) (b) (c)

Figure 3: Heterogeneous nucleation of new crystallites (a) and generation of voids (b) at the interface between metal substrate and transparent overlayer. The images from this study were used for cover art of an issue of the Journal of Applied Physics (c).

E. T. Karim, M. Shugaev, C. Wu, Z. Lin, R. F. Hainsey, and L. V. Zhigilei, *J. Appl. Phys.* **115**, 183501, 2014.

E. T. Karim, M. V. Shugaev, C. Wu, Z. Lin, H. Matsumoto, M. Conneran, J. Kleinert, R. F. Hainsey, and L. V. Zhigilei, *Appl. Phys. A*, in press.

(4) *Microscopic mechanisms of laser spallation and ablation of metal targets from large-scale molecular dynamics simulations.* The microscopic mechanisms of femtosecond laser ablation of an Al target are investigated in large-scale massively parallel atomistic simulations performed with a computational model combining classical molecular dynamics technique with a continuum description of the laser excitation and subsequent relaxation of conduction band electrons. The relatively large lateral size of the computational systems used in the simulations enables a detailed analysis of the evolution of multiple voids generated in a sub-surface region of the irradiated target in the spallation regime, when the material ejection is driven by the

relaxation of laser-induced stresses. The nucleation, growth, and coalescence of voids take place within a broad (~100 nm) region of the target, leading to the formation of a transient foamy structure of interconnected liquid regions and eventual separation (or spallation) of a thin liquid layer from the bulk of the target. The thickness of the spalled layer is decreasing from the maximum of ~50 nm while the temperature and ejection velocity are increasing with increasing fluence. At a fluence of ~2.5 times the spallation threshold, the top part of the target reaches the conditions for an explosive decomposition into vapor and small clusters/droplets, marking the transition to the phase explosion regime of laser ablation. This transition is signified by a change in the composition of the ablation plume from large liquid droplets to a mixture of vapor-phase atoms and clusters/droplets of different sizes. The clusters of different sizes are spatially segregated in the expanding ablation plume, where small/medium size clusters present in the middle of the plume are followed by slower (velocities of less than 3 km/s) large droplets consisting of more than 10,000 atoms.

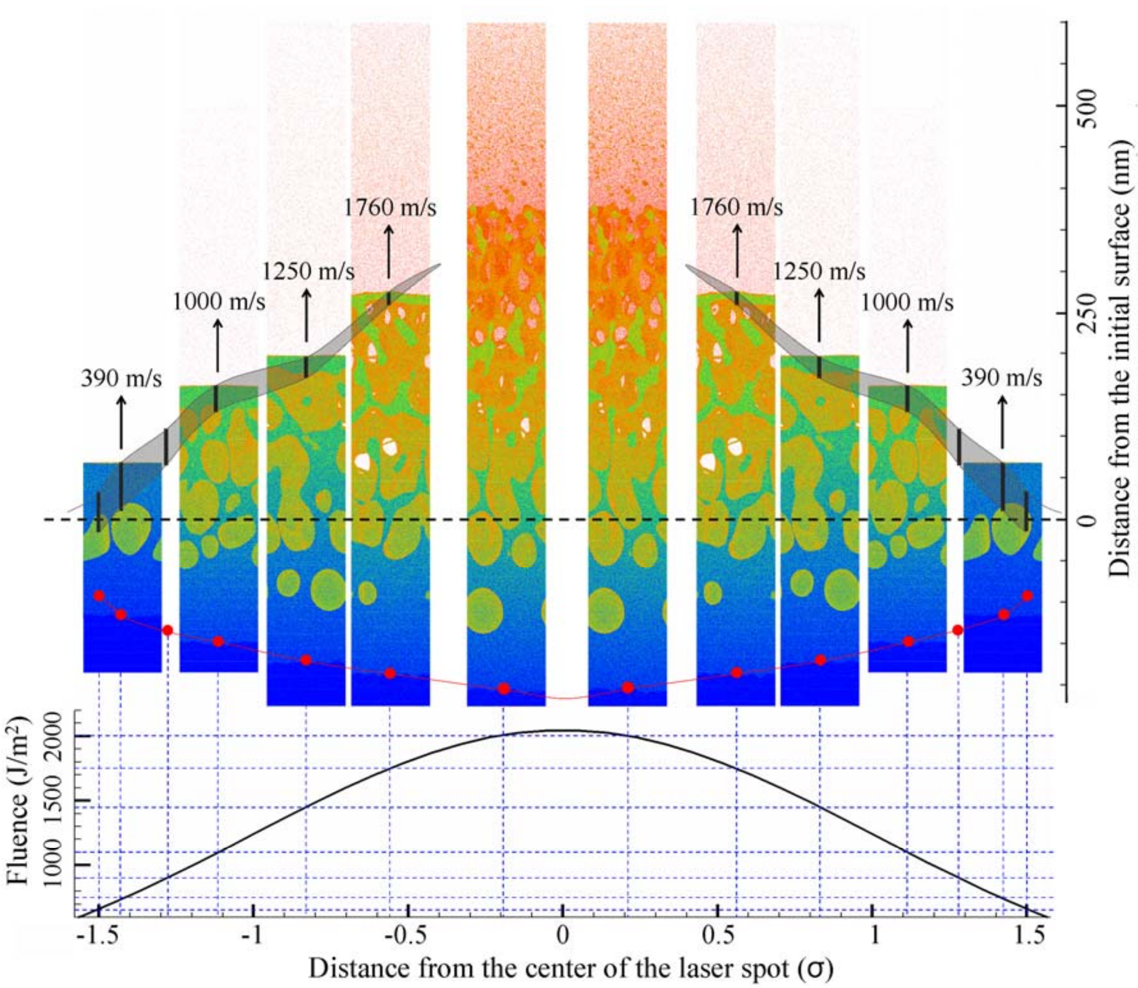


Figure 4: An illustration of the "mosaic" approach to mapping the results of atomistic simulations to processes occurring at the scale of the whole laser spot. Snapshots from the atomistic simulations of Al target irradiated by 100 fs laser pulses at different local fluences are shown for 150 ps after the irradiation.

C. Wu and L. V. Zhigilei, *Appl. Phys. A* **114**, 11-32, 2014.

(5) ***Investigation of surface nanostructuring of metal surfaces by short laser pulses***. Structural transformations in a shallow surface regions of bulk metal targets irradiated by femtosecond laser pulses are investigated in large-scale atomistic simulations. The simulations reveal a complex interplay of fast laser melting, rapid resolidification, and dynamic relaxation of laser-induced stresses that leads to the formation of a subsurface porous region covered by a nanocrystalline surface layer. The generation of the porous region is consistent with the experimental observation of surface "swelling" occurring at laser fluences below the spallation/ablation threshold and may be related to the incubation effect in multipulse laser ablation of metals. The nanocrystalline layer is produced by massive nucleation of crystallites triggered by a deep undercooling of the melted surface region experiencing fast quenching at a rate on the order of 10^{11} K/s. The predicted surface structure features random crystallographic orientation of nanograins and a high density of stacking faults, twins, and nanoscale twinned structural elements with fivefold symmetry, which suggests high hardness and possible enhancement of catalytic activity of the surface. At a higher laser fluence, above the spallation threshold, the generation of frozen nanospikes with an unusual structure represented by a continuous network of pentagonal twinned structural elements arranged into a polyicosahedral structure is observed.

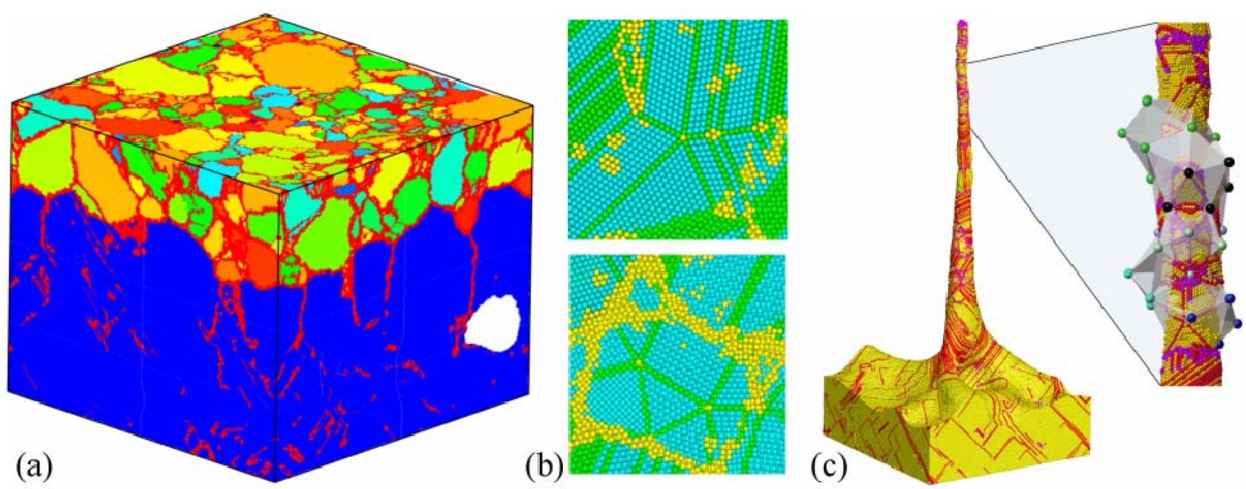


Figure 5: Results of atomistic simulations of 100 fs laser irradiation of Ag targets at absorbed laser fluences of 850 J/m^2 (a,b) and 900 J/m^2 (c). The simulations predict formation of nanocrystalline surface structures (a) with high density of stacking faults and twins (b), as well as a nanospike featuring icosahedral nanostructures (c).

C. Wu, M. S. Christensen, J.-M. Savolainen, P. Balling, and L. V. Zhigilei, *Phys. Rev. B* **91**, 035413, 2015.

C. Wu and L. V. Zhigilei, J. Phys. Chem. C, in press, 2016, DOI: 10.1021/acs.jpcc.6b00013

(6) ***Carbon surface oxidation in a reacting boundary layer with detailed kinetics:*** Complex interaction between homogeneous gas-phase kinetics and heterogeneous surface kinetics over a reacting carbon surface was investigated by considering a surface heated by a continuous CO_2-laser source. The overall reactivity of the surface was parameterized by free-stream velocity, surface temperature due to laser heating, and the porosity of the material. The companion

experiments were performed at AFIT by Dr. Perram's group. Figure 6 shows the computational domain implemented with the heated carbon sample highlighted by red, while Figure 7 shows a typical contour plot of major species over a non-porous carbon surface with a density of ~2.2 gm/cm^3. Figure 7a corresponds to a low laser fluence case with the sample surface temperature held constant at 1800 K and Figure 7 corresponds to a high temperature case of 2600 K. The temperature dependence can be related to the large activation energy of key carbon oxidation reactions, $C_s+1/2O_2 \rightarrow CO$ (R1), $C_s + CO_2 \rightarrow 2CO$ (R2), $C_s+H_2O \rightarrow CO+H_2$ (R3), while the flow velocity influences the overall reactivity due to transport of species across reacting boundary layer and time available for chemical reactions, i.e. finite Damkohler number (ratio of diffusion time to chemical reaction time) effect. The sample porosity affects the number of active sites available in thin porous layer of the sample at the interface between the gas and the sold, which is accounted in the pre-exponential collision frequency term of the semi-global surface reaction expressions.

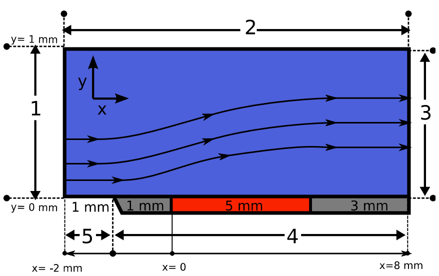


Figure 6: Computational domain showing a flat surface with a 5mm imbedded carbon sample under investigation.

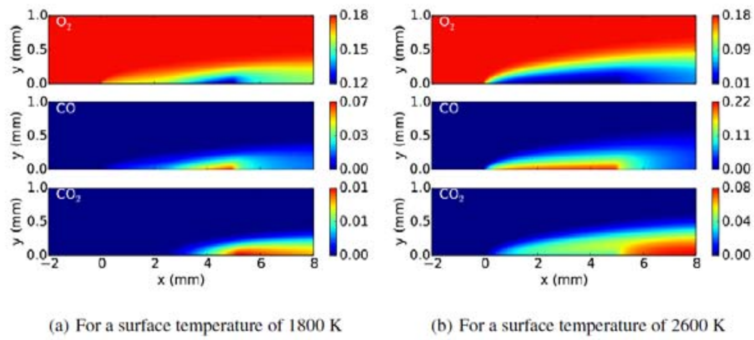


Figure 7: Contour plots of O_2, CO, CO_2 for a non-porous carbon surface heated to two surface temperatures of 1800 and 2600 K, respectively, in a reacting boundary layer with a free-stream velocity of 30 m/s.

The CO formed at the surface by reactions R1-R3 diffuses to the gas-phase and undergoes homogeneous oxidation. The latter depends on the availability of OH (wet CO oxidation chemistry) and also depends strongly on the flow residence time and surface temperature. The low Damkohler conditions at the leading edge lead of the sample leads to a homogeneous reaction kernel while in the downstream region, a diffusion controlled reaction front is established. Such differences may manifest as an attached CO flame or detached CO flame, as shown in Figure 8. The hyper-spectral image studies at AFIT was focused on observing these subtle reacting flow structure variations by measuring the key species CO, O_2, CO_2, H_2O, etc. with sufficient resolution.

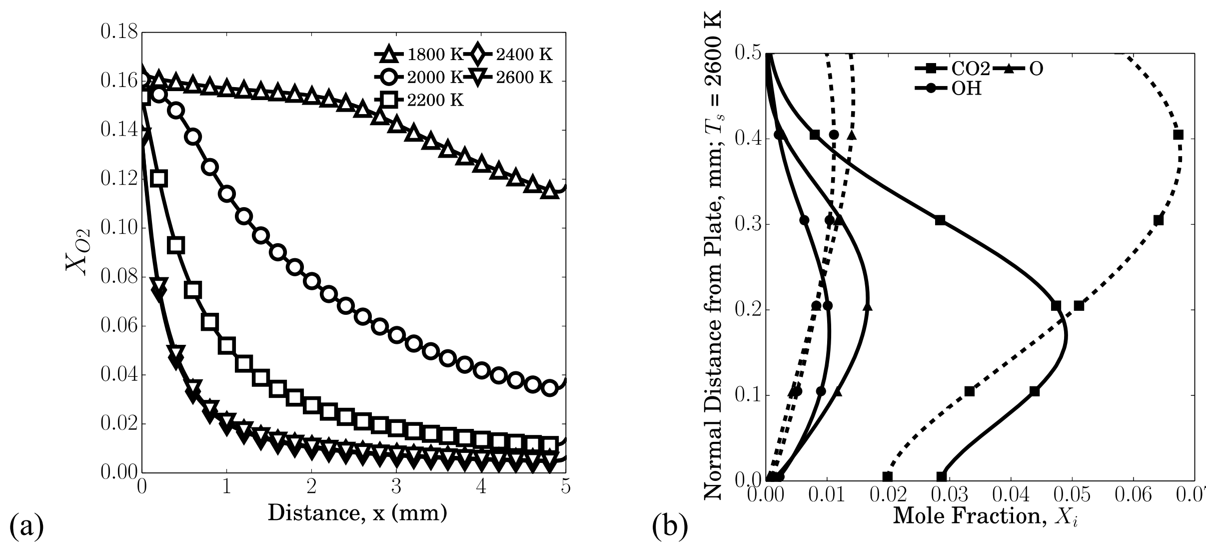
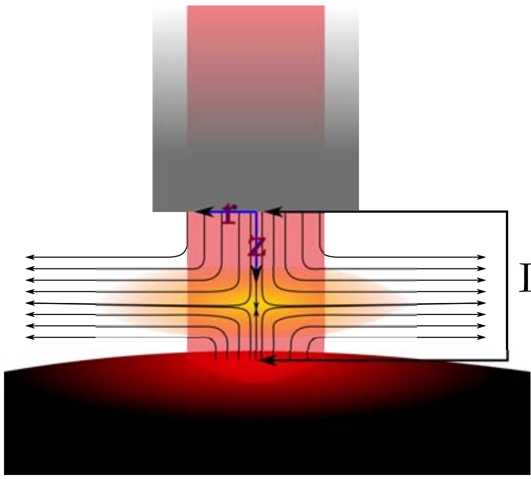


Figure 8: (a) Oxygen mole fraction variation along the surface as a function surface temperature variation for non-porous carbon surface and for a flow velocity of 30 m/s, and (b) the reacting layer structure normal to the surface showing a detached CO oxidation region forming peak CO2 away from the surface at 2600 K. Importance of $Cs+CO_2$ reaction on CO_2, OH, O-atoms is shown in (b) by including (dashed lines) and by excluding (solid lines) this reaction in the surface chemistry model.

R. F. Johnson and H. K. Chelliah, accepted for publication in *Combustion Theory and Modeling*, 2016, in print.

(7) ***Carbon surface oxidation in a stagnating-point flow field with detailed kinetics:*** Besides ablation/oxidation of carbon surfaces described in section 6 in reacting boundary layers with the free-stream velocity parallel to the surface, another useful configuration is the reaction occurring in a stagnating-point flow filed. This configuration may correspond to the nose of a missile as illustrated in Figure 9. In this configuration, the detached CO-oxidation flame is a strong function of the flow strain effects, which can also be characterized by the Damkohler number. Parametric investigations were performed in this configuration to support the experiments being performed at AFIT to obtain fundamental data on the structure due to coupled heterogeneous-homogeneous reaction system. One key objective was to explore the ideal surface temperature and moisture content that yields the maximum CO_2 layer thickness that is suitable for hyper-spectral imaging investigations, as shown in Figure 10.



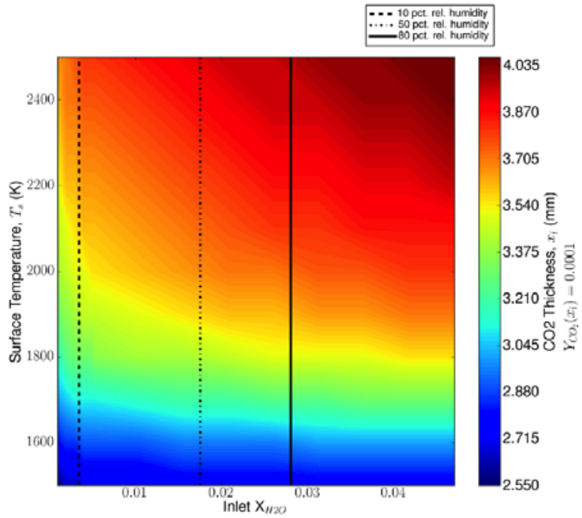


Figure 9: Stagnation-point flow configuration corresponding to the nose region of missile heated with a high-energy laser source.

Figure 10: Parametric investigation of reacting CO2 layer thickness amenable for hyper-spectral imaging as a function of surface temperature and moisture content.

Unlike the parallel reacting boundary layer flow filed considered in section (6), the present stagnation-point flow offers much finer control of the interaction between the homogeneous and heterogeneous chemistry as well as experimental exploration of the flame structure. Figure 11 shows the predicted CO_2 mole fraction contour plot indicating the stabilization of a detached CO_2 layer over the laser heated carbon surface, while Figure 12 shows a line plot of major species normal to the surface. Experimental quantification of this flame structure can provide the first ever detailed validation of the complex interaction between semi-global heterogeneous chemical kinetic models and detailed homogeneous CO-oxidation models assuming that the hyper-spectral imaging camera at AFIT can resolve reaction layer thickness of the order of 3 mm.

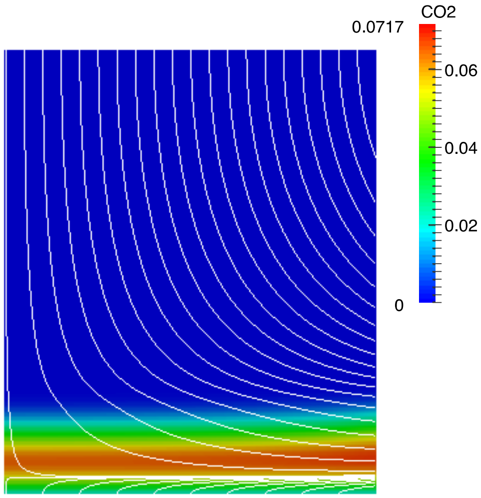


Figure 11: Predicted CO_2 contour plot in stagnation-point flow field (only one-half is shown) indicating the establishment of a lifted CO_2 flame.

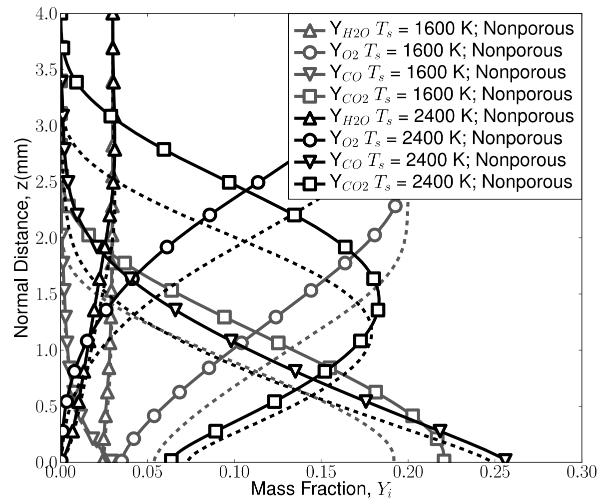


Figure 12: Parametric effects of surface temperature on the reacting flame structure over a non-porous carbon surface subjected to a global flow strain rate of 500 s^{-1}. Solid lines are along the axis of symmetry (r = 0) and dashed lines are at r = 10 mm showing a nearly planar flame structure.

The counterflow stagnating-point flow geometry was also used to validate the computational approach adopted by considering the details of the reacting flow structure of a well-known hydrogen chemistry model. As part of this investigation, the effects of nozzle diameter and nozzle separation distance from the stagnation plane, a nagging fundamental issue was addressed in great detailed as part of this work, and is expected to have a significant and lasting impact in the combustion field.

R. F. Johnson and H. K. Chelliah, Coupled heterogeneous and homogeneous reactions in a two-dimensional stagnation-point flow field, to be submitted *Combustion Theory and Modeling,* 2016.

R. F. Johnson, A. C. VanDine, G. Esposito, H. K. Chelliah, On the axisymmetric counterflow flame simulations: Is there an optimal nozzle diameter and separation distance to apply quasi one-dimensional theory?, *Combustion Science and Technology* **187**, 37-59, 2015.

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Leonid Zhigilei

Program Manager
The AFOSR Program Manager currently assigned to the award

Jason Marshall

Reporting Period Start Date

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Abstract

The overall goal of the project was to develop advanced computational models for analysis of the effect of external flow on material removal in laser ablation of metals in oxidizing environment and under the influence of air flow. The models are applied for investigation of the combined effect of the laser energy deposition, chemical reactions, and air flow on the material removal rate and the nature of laser damage/modification of irradiated targets. The results of this project include the elucidation of (1) the relative contributions of the recoil vapor pressure and Marangoni effects on the melt dynamics and melt-through time in the laser melting of free-standing aluminum films, (2) the effect of the external gas pressure on the flow structures and the mechanisms of the alumina and oxygen transport to the target surface, (3) the distinct characteristics of short pulse laser interactions with a metal target under conditions of spatial confinement by a solid transparent overlayer, (4) the conditions for the formation of nanocrystalline surface layer, sub-surface voids, and frozen surface nanospikes in surface nanostructuring by short laser pulses, (5) the effect of sample porosity on laser-induced carbon pyrolysis and oxidation.

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4. E. T. Karim, M. Shugaev, C. Wu, Z. Lin, R. F. Hainsey, and L. V. Zhigilei, Atomistic simulation study of short pulse laser interactions with a metal target under conditions of spatial confinement by a transparent overlayer, J. Appl. Phys. 115, 183501, 2014.

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11. R. F. Johnson and H. K. Chelliah, Laminar reactive boundary layer simulation of an ablating heated carbon surface, 51st AIAA Aerospace Sciences Meeting including the New Horizons Forum and Aerospace Exposition, 184, 2013.

12. R. F. Johnson, A. C. VanDine, G. Esposito, and H. K. Chelliah, On the axisymmetric counterflow flame simulations: Is there an optimal nozzle diameter and separation distance to apply quasi one-dimensional theory?, Combustion Science and Technology 187, 37-59, 2015.

13. R. F. Johnson and H. K. Chelliah, Numerical simulation of two-dimensional flow over a heated carbon surface with coupled heterogeneous and homogeneous reactions, accepted for publication in Combustion Theory and Modeling, 2016, in print.

14. R. F. Johnson and H. K. Chelliah, Coupled heterogeneous and homogeneous reactions in a two-dimensional stagnation-point flow field, to be submitted Combustion Theory and Modeling, 2016.

Changes in research objectives (if any):

Change in AFOSR Program Manager, if any:

Extensions granted or milestones slipped, if any:

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LRIR Title

Reporting Period

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Research Objectives

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